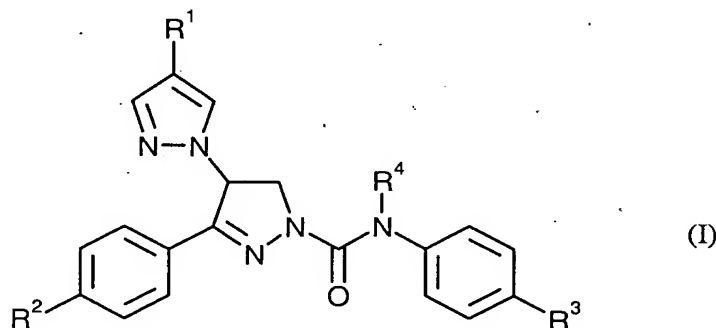


Substituted pyrazolinecarboxanilides

The present invention relates to novel substituted pyrazolinecarboxanilides, to processes for their preparation and to their use as pesticides, in particular as arthropodicides.

It is known that certain substituted pyrazolinecarboxanilides have insecticidal and acaricidal properties (cf., for example, EP-A 0 438 690, EP-A 0 679 644). However, the activity of these compounds is, in particular at low concentration of active compounds and low application rates, not always entirely satisfactory.

This invention provides novel substituted pyrazolinecarboxanilides of the formula (I)



in which

R¹ represents halogen,

R² represents cyano, halogen, haloalkyl, haloalkoxy, alkylthio, haloalkylthio, alkylsulphinyl, haloalkylsulphinyl, alkylsulphonyl or haloalkylsulphonyl,

R³ represents cyano, halogen, haloalkyl, haloalkoxy, alkylthio, haloalkylthio, alkylsulphinyl, haloalkylsulphinyl, alkylsulphonyl or haloalkylsulphonyl and

R⁴ represents cyanoalkyl.

Depending, if appropriate, on the type and number of substituents, the compounds of the formula (I) can be present as geometrical and/or optical isomers, regioisomers and/or configurational isomers or isomer mixtures thereof of varying composition. What is claimed by the invention are both the pure isomers and the isomer mixtures.

Furthermore, it has been found that the substituted pyrazolinecarboxanilides of the formula

(I) are obtained when

(a) substituted pyrazolines of the formula (II)

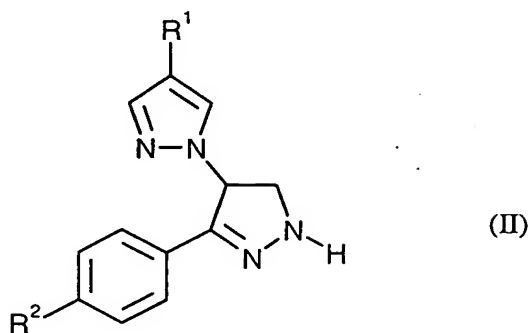
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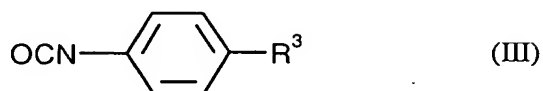
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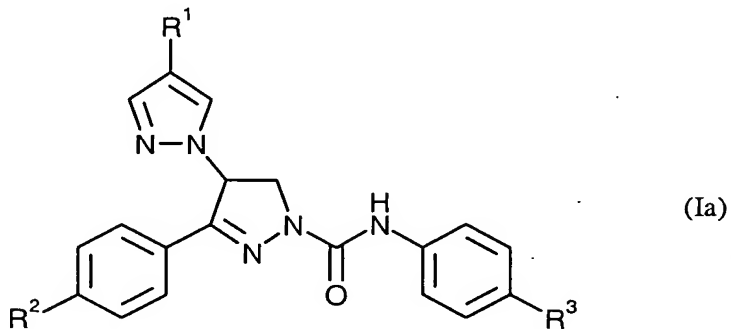
in which R¹ and R² are as defined above,

are reacted with isocyanates of the formula (III)



in which R³ is as defined above,

if appropriate in the presence of a diluent and if appropriate in the presence of a catalyst, and the resulting substituted pyrazolinecarboxanilides of the formula (Ia)



in which

R¹, R² and R³ are as defined above,

are reacted with halogen compounds of the formula (IV)



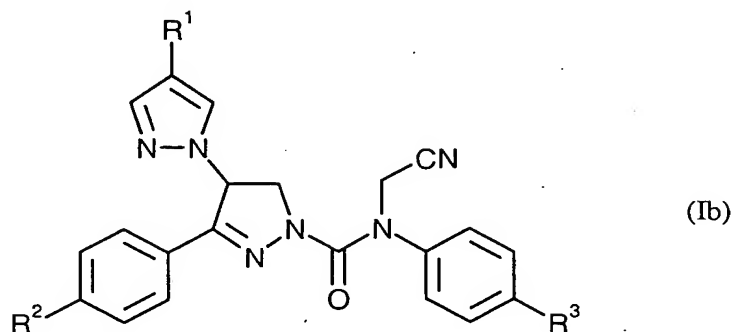
in which

R⁴ is as defined above and

X¹ represents halogen,

if appropriate in the presence of a diluent and if appropriate in the presence of a base,

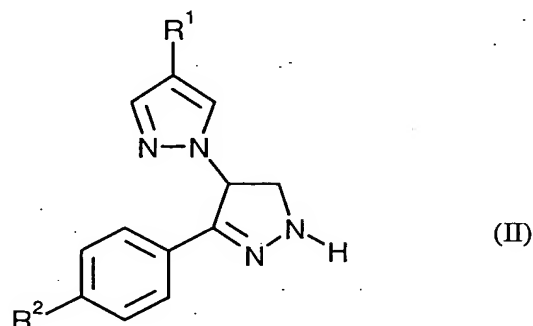
or that the substituted pyrazolinecarboxanilides of the formula (Ib)



in which R^1 , R^2 and R^3 are as defined above,

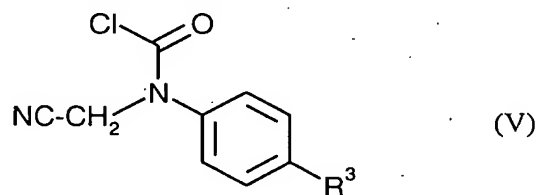
5 are obtained by

(b) reacting substituted pyrazolines of the formula (II)



in which R^1 and R^2 are as defined above,

10 with carbamoyl chlorides of the formula (V)



in which R^3 is as defined above,

in the presence of a diluent and if appropriate in the presence of a base.

15

Finally, it has been found that the novel substituted pyrazolinecarboxanilides of the formula (I) have strongly pronounced biological properties and are suitable especially for controlling animal pests, in particular insects, arachnids and nematodes, encountered in agriculture, in forests, in the protection of stored products and materials and in the hygiene sector.

The formula (I) provides a general definition of the substituted pyrazolinecarboxanilides according to the invention.

5 Preferred substituents or ranges of radicals listed in the formulae mentioned above and below are illustrated below:

R¹ preferably represents chlorine, bromine or iodine.

R¹ particularly preferably represents chlorine or bromine.

10

R² preferably represents cyano, fluorine, chlorine, bromine, iodine, C₁-C₄-haloalkyl, C₁-C₄-haloalkoxy, C₁-C₄-alkylthio, C₁-C₄-haloalkylthio, C₁-C₄-alkylsulphinyl, C₁-C₄-haloalkylsulphinyl, C₁-C₄-alkylsulphonyl or C₁-C₄-haloalkylsulphonyl.

15

R² particularly preferably represents cyano, fluorine, chlorine, bromine, iodine, monofluoromethyl, difluoromethyl, trifluoromethyl, monochloromethyl, dichloromethyl, trichloromethyl, chlorodifluoromethyl, fluorodichloromethyl, monofluoroethyl, difluoroethyl, trifluoroethyl, tetrafluoroethyl, pentafluoroethyl, monochloroethyl, dichloroethyl, trichloroethyl, chlorofluoroethyl, chlorodifluoroethyl, chlorotrifluoroethyl, fluorodichloroethyl, dichlorodifluoroethyl, difluoromethoxy, trifluoromethoxy, chlorodifluoromethoxy, fluoroethoxy, difluoroethoxy, trifluoroethoxy, tetrafluoroethoxy, chloroethoxy, dichloroethoxy, chlorofluoroethoxy, methylthio, ethylthio, n- or i-propylthio, difluoromethylthio, trifluoromethylthio, chlorodifluoromethylthio, methylsulphinyl, ethylsulphinyl, difluoromethylsulphinyl, trifluoromethylsulphinyl, methylsulphonyl, ethylsulphonyl, difluoromethylsulphonyl or trifluoromethylsulphonyl.

20

25

R² very particularly preferably represents cyano, fluorine, chlorine, bromine, iodine, difluoromethyl, trifluoromethyl, dichloromethyl, trichloromethyl, chlorodifluoromethyl, fluorodichloromethyl, difluoromethoxy, trifluoromethoxy, chlorodifluoromethoxy, difluoromethylthio, trifluoromethylthio, chlorodifluoromethylthio, trifluoromethylsulphinyl or trifluoromethylsulphonyl.

30

R³ preferably represents cyano, fluorine, chlorine, bromine, iodine, C₁-C₄-haloalkyl, C₁-C₄-haloalkoxy, C₁-C₄-alkylthio, C₁-C₄-haloalkylthio, C₁-C₄-alkylsulphinyl, C₁-C₄-haloalkylsulphinyl, C₁-C₄-alkylsulphonyl or C₁-C₄-haloalkylsulphonyl.

35

R³ particularly preferably represents cyano, fluorine, chlorine, bromine, iodine, monofluoromethyl, difluoromethyl, trifluoromethyl, monochloromethyl, dichloro-

5 methyl, trichloromethyl, chlorodifluoromethyl, fluorodichloromethyl, monofluoroethyl, difluoroethyl, trifluoroethyl, tetrafluoroethyl, pentafluoroethyl, monochloroethyl, dichloroethyl, trichloroethyl, chlorofluoroethyl, chlorodifluoroethyl, chlorotrifluoroethyl, fluorodichloroethyl, dichlorodifluoroethyl, difluoromethoxy, trifluoromethoxy, chlorodifluoromethoxy, fluoroethoxy, difluoroethoxy, trifluoroethoxy, tetrafluoroethoxy, chloroethoxy, dichloroethoxy, chlorofluoroethoxy, methylthio, ethylthio, n- or i-propylthio, difluoromethylthio, trifluoromethylthio, chlorodifluoromethylthio, methylsulphinyl, ethylsulphinyl, difluoromethylsulphinyl, trifluoromethylsulphinyl, methylsulphonyl, ethylsulphonyl, difluoromethylsulphonyl or trifluoromethylsulphonyl.

10 R^3 very particularly preferably represents cyano, fluorine, chlorine, bromine, iodine, difluoromethyl, trifluoromethyl, dichloromethyl, trichloromethyl, chlorodifluoromethyl, fluorodichloromethyl, difluoromethoxy, trifluoromethoxy, chlorodifluoromethoxy, difluoromethylthio, trifluoromethylthio, chlorodifluoromethylthio, trifluoromethylsulphinyl or trifluoromethylsulphonyl.

15 R^4 preferably represents cyano- C_1 - C_4 -alkyl.

R^4 particularly preferably represents cyanomethyl, cyanoethyl or cyanopropyl.

R^4 very particularly preferably represents cyanomethyl.

20 Especially preferred are compounds of the formula (I) in which R^1 represents chlorine.

Especially preferred are compounds of the formula (I) in which R^4 represents cyanomethyl.

Especially preferred are compounds of the formula (I) in which R^1 represents chlorine and R^4 represents cyanomethyl.

25 Especially preferred are compounds of the formula (I) in which R^2 represents fluorine, chlorine or bromine, particularly preferably chlorine.

Especially preferred are compounds of the formula (I) in which R^3 represents cyano, chlorine, bromine, trifluoromethyl, trifluoromethoxy or trifluoromethylthio.

30 The general or preferred radical definitions or illustrations listed above apply both to the end products and, correspondingly, to the starting materials and intermediates. These radical definitions can be combined with one another as desired, i.e. including combinations between the respective preferred ranges.

35 Preference according to the invention is given to the compounds of the formula (I) which contain a combination of the meanings listed above as being preferred.

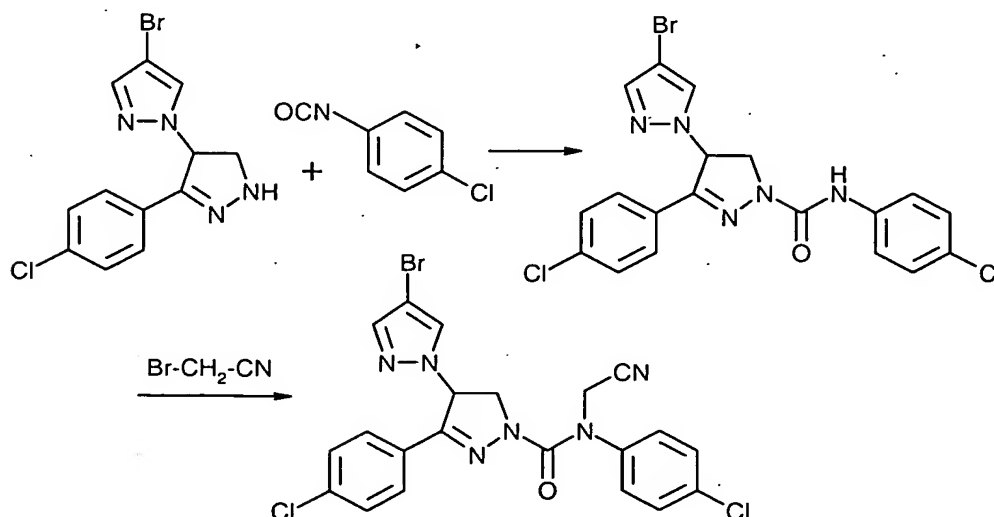
Particular preference according to the invention is given to the compounds of the formula (I) which contain a combination of the meanings listed above as being particularly preferred.

Very particular preference according to the invention is given to the compounds of the formula (I) which contain a combination of the meanings listed above as being very particularly preferred.

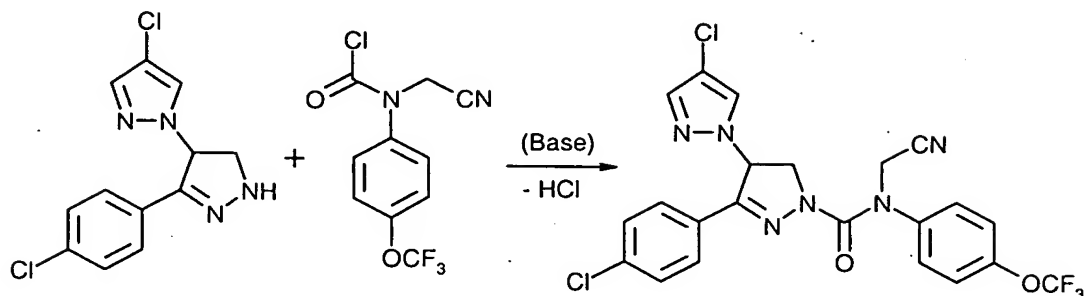
In the radical definitions given above and below, carbon radicals, such as alkyl, are in each case straight-chain or branched as far as this is possible – including in combination with hetero atoms such as alkoxy.

Halogen-substituted radicals, such as, for example, haloalkyl, are mono- or polyhalogenated. In the case of polyhalogenation, the halogen atoms can be identical or different. Here, halogen represents fluorine, chlorine, bromine and iodine, in particular fluorine, chlorine and bromine.

Using, for example, 3-(4-chlorophenyl)-4-(4-bromopyrazol-1-yl)-4,5-dihydro-1H-pyrazole and 4-chlorophenyl isocyanate and in the subsequent step bromoacetonitrile as starting materials, the course of the reaction of the process (a) according to the invention can be represented by the formula scheme below:



Using, for example, 3-(4-chlorophenyl)-4-(4-chloropyrazol-1-yl)-4,5-dihydro-1H-pyrazole and N-cyanomethyl-N-(4-trifluoromethoxyphenyl)carbamoyl chloride as starting materials, the course of the reaction of the process (b) according to the invention can be represented by the formula scheme below:



Formula (II) provides a general definition of the substituted pyrazolines to be used as starting materials for carrying out the processes (a) and (b) according to the invention. In this formula, R¹ and R² preferably, particularly preferably and very particularly preferably have those meanings which have already been mentioned in connection with the description of the compounds of the formula (I) according to the invention as being preferred, particularly preferred and very particularly preferred, respectively, for these radicals.

The substituted pyrazolines of the formula (II) are known and/or can be prepared by processes known per se (cf. EP-A 0 438 690, EP-A 0 546 420 and the Preparation Examples).

The formula (III) provides a general definition of the isocyanates further to be used as starting materials in the process (a) according to the invention. In this formula, R³ preferably, particularly preferably and very particularly preferably has those meanings which have already been mentioned in connection with the description of the compounds of the formula (I) according to the invention as being preferred, particularly preferred and very particularly preferred, respectively, for this radical.

The isocyanates of the formula (III) are generally known compounds of organic chemistry and/or can be obtained in a generally known manner.

The formula (IV) provides a general definition of the halogen compounds further to be used as starting materials for carrying out the process (a) according to the invention. In this formula, R⁴ preferably, particularly preferably and very particularly preferably has those meanings which have already been mentioned in connection with the description of the compounds of the formula (I) according to the invention as being preferred, particularly preferred and very particularly preferred, respectively, for this radical. X¹ preferably represents chlorine or bromine.

The halogen compounds of the formula (IV) are generally known compounds of organic chemistry.

5 Formula (V) provides a general definition of the carbamoyl chlorides further to be used as starting materials in the process (b) according to the invention. In this formula, R^3 preferably, particularly preferably and very particularly preferably has those meanings which have already been mentioned in connection with the description of the compounds of the formula (I) according to the invention as being preferred, particularly preferred and very
10 particularly preferred, respectively, for this radical.

The carbamoyl chlorides of the formula (V) are known and/or can be prepared by processes known per se (cf. DE-A 27 30 325, DE-A 101 35 551 dated 20 July 2001 and the Preparation Examples).

15 Process (a) according to the invention is preferably carried out using one or more diluents. Suitable diluents are virtually all inert inorganic solvents. These preferably include aliphatic and aromatic, optionally halogenated hydrocarbons, such as pentane, hexane, heptane, cyclohexane, petroleum ether, benzine, ligroine, benzene, toluene, xylene, methylene
20 chloride, ethylene chloride, chloroform, carbon tetrachloride, chlorobenzene and *o*-dichlorobenzene, ethers, such as diethyl ether, diisopropyl ether, diisobutyl ether, methyl tert-butyl ether, methyl tert-amyl ether, glycol dimethyl ether and diglycol dimethyl ether, tetrahydrofuran and dioxane, ketones, such as acetone, methyl ethyl ketone, ethyl isopropyl ketone or methyl isobutyl ketone, esters, such as methyl acetate or ethyl acetate, nitriles,
25 such as acetonitrile or propionitrile, amides, such as N,N-dimethylformamide, N,N-dimethylacetamide and N-methylpyrrolidone, and also dimethyl sulphoxide, tetramethylene sulphone or hexamethylphosphoric triamide.

30 Process (a) according to the invention is preferably also carried out using a catalyst. Suitable catalysts are in particular tertiary organic amines, such as, for example, triethylamine.

Suitable bases for carrying out the processes (a) and (b) according to the invention are all acid binders customarily used for such reactions. Preference is given to using alkali metal and alkaline earth metal hydrides, such as lithium hydride, sodium hydride, potassium
35 hydride or calcium hydride; alkaline earth metal and alkali metal hydroxides, such as lithium hydroxide, sodium hydroxide, potassium hydroxide or calcium hydroxide; alkali metal and

alkaline earth metal carbonates or bicarbonates, such as sodium carbonate or potassium carbonate or sodium bicarbonate or potassium bicarbonate or calcium carbonate; alkali metal acetates, such as sodium acetate or potassium acetate, alkali metal alkoxides, such as sodium tert-butoxide or potassium tert-butoxide; furthermore basic nitrogen compounds, such as trimethylamine, triethylamine, tripropylamine, tributylamine, diisobutylamine, dicyclohexylamine, ethyldiisopropylamine, ethyldicyclohexylamine, N,N-dimethylbenzylamine, N,N-dimethylaniline, pyridine, 2-methyl-, 3-methyl-, 4-methyl-, 2,4-dimethyl-, 2,6-dimethyl-, 2-ethyl-, 4-ethyl- and 5-ethyl-2-methylpyridine, 1,5-diazabicyclo[4.3.0]non-5-ene (DBN), 1,8-diazabicyclo[5.4.0]undec-7-ene (DBU), 1,4-diazabicyclo[2.2.2]octane (DABCO).

In the process (a) according to the invention, the reaction temperatures can be varied within a relatively wide range. In general, the process is carried out at temperatures between 0°C and 120°C, preferably at temperatures between 20°C and 80°C.

The process (a) according to the invention is generally carried out under atmospheric pressure. However, it is also possible to operate under elevated or reduced pressure.

For carrying out the process (a) according to the invention, the starting materials are generally employed in approximately equimolar amounts. However, it is also possible to use a relatively small excess of one of the two components employed. Work-up is carried out by customary methods (cf. the Preparation Examples).

Process (b) according to the invention is preferably carried out using one or more diluents. Suitable diluents are virtually all inert organic solvents. These preferably include aliphatic and aromatic, optionally halogenated hydrocarbons, such as pentane, hexane, heptane, cyclohexane, petroleum ether, benzine, logroine, benzene, toluene, xylene, methylene chloride, ethylene chloride, chloroform, carbon tetrachloride, chlorobenzene and o-dichlorobenzene, ethers, such as diethyl ether, diisopropyl ether, di-tert-butyl ether, glycol dimethyl ether and diglycol dimethyl ether, tetrahydrofuran and dioxane, ketones, such as acetone, methyl ethyl ketone, methyl isopropyl ketone or methyl isobutyl ketone, esters, such as methyl acetate or ethyl acetate, nitriles, such as, for example, acetonitrile or propionitrile, amides, such as, for example, N,N-dimethylformamide, N,N-dimethylacetamide and N-methylpyrrolidone, and also dimethyl sulphoxide, tetramethylene sulphone or hexamethylphosphoric triamide.

In the process (b) according to the invention, the reaction temperatures can be varied within a relatively wide range. In general, the process is carried out at temperatures between -20°C and 120°C, preferably at temperatures between 0°C and 80°C.

5 The process (b) according to the invention is generally carried out at atmospheric pressure. However, it is also possible to operate under elevated or reduced pressure.

For carrying out the process (b) according to the invention, the starting materials are generally employed in approximately equimolar amounts. However, it is also possible to use
10 an excess of halide and base. Work-up is carried out by customary methods (cf. the Preparation Examples).

The active compounds are suitable for protecting plants and plant organs, for increasing the harvest yields, for improving the quality of the harvested material and for controlling animal
15 pests, in particular insects, arachnids and nematodes, which are encountered in agriculture, in forests, in gardens and leisure facilities, in the protection of stored products and of materials, and in the hygiene sector, and have good plant tolerance and favourable toxicity to warm-blooded animals and are tolerated well by the environment. They may be preferably employed as plant protection agents. They are active against normally sensitive and
20 resistance species and against all or some stages of development. The abovementioned pests include:

From the order of the Isopoda, for example, *Oniscus asellus*, *Armadillidium vulgare* and *Porcellio scaber*.

25 From the order of the Diplopoda, for example, *Blaniulus guttulatus*.

From the order of the Chilopoda, for example, *Geophilus carpophagus* and *Scutigera* spp.

From the order of the Symphyla, for example, *Scutigera* spp.

From the order of the Thysanura, for example, *Lepisma saccharina*.

From the order of the Collembola, for example, *Onychiurus armatus*.

30 From the order of the Orthoptera, for example, *Acheta domesticus*, *Grylotalpa* spp., *Locusta migratoria migratorioides*, *Melanoplus* spp. and *Schistocerca gregaria*.

From the order of the Blattaria, for example, *Blatta orientalis*, *Periplaneta americana*, *Leucophaea maderae*, *Blattella germanica*.

From the order of the Dermaptera, for example, *Forficula auricularia*.

35 From the order of the Isoptera, for example, *Reticulitermes* spp.

From the order of the Phthiraptera, for example, *Pediculus humanus corporis*, *Haematopinus* spp., *Linognathus* spp., *Trichodectes* spp. and *Damalinia* spp.

From the order of the Thysanoptera, for example, *Hercinothrips femoralis*, *Thrips tabaci*, *Thrips palmi* and *Frankliniella occidentalis*.

- 5 From the order of the Heteroptera, for example, *Eurygaster* spp., *Dysdercus intermedius*, *Piesma quadrata*, *Cimex lectularius*, *Rhodnius prolixus* and *Triatoma* spp.

- From the order of the Homoptera, for example, *Aleurodes brassicae*, *Bemisia tabaci*, *Trialeurodes vaporariorum*, *Aphis gossypii*, *Brevicoryne brassicae*, *Cryptomyzus ribis*, *Aphis fabae*, *Aphis pomi*, *Eriosoma lanigerum*, *Hyalopterus arundinis*, *Phylloxera vastatrix*,
10 *Pemphigus* spp., *Macrosiphum avenae*, *Myzus* spp., *Phorodon humuli*, *Rhopalosiphum padi*, *Empoasca* spp., *Euscelis bilobatus*, *Nephotettix cincticeps*, *Lecanium corni*, *Saissetia oleae*, *Laodelphax striatellus*, *Nilaparvata lugens*, *Aonidiella aurantii*, *Aspidiotus hederae*, *Pseudococcus* spp. and *Psylla* spp.

- From the order of the Lepidoptera, for example, *Pectinophora gossypiella*, *Bupalus piniarius*,
15 *Cheimatobia brumata*, *Lithocolletis blancardella*, *Hyponomeuta padella*, *Plutella xylostella*, *Malacosoma neustria*, *Euproctis chrysorrhoea*, *Lymantria* spp., *Bucculatrix thurberiella*, *Phyllocnistis citrella*, *Agrotis* spp., *Euxoa* spp., *Feltia* spp., *Earias insulana*, *Heliothis* spp., *Mamestra brassicae*, *Panolis flammea*, *Spodoptera* spp., *Trichoplusia ni*, *Carpocapsa pomonella*, *Pieris* spp., *Chilo* spp., *Pyrausta nubilalis*, *Ephestia kuehniella*, *Galleria mellonella*,
20 *Tineola bisselliella*, *Tinea pellionella*, *Hofmannophila pseudospretella*, *Cacoecia podana*, *Capua reticulana*, *Choristoneura fumiferana*, *Clysia ambiguella*, *Homona magnanima*, *Tortrix viridana*, *Cnaphalocerus* spp., *Oulema oryzae*.

- From the order of the Coleoptera, for example, *Anobium punctatum*, *Rhizopertha dominica*,
25 *Bruchidius obtectus*, *Acanthoscelides obtectus*, *Hylotrupes bajulus*, *Agelastica alni*, *Leptinotarsa decemlineata*, *Phaedon cochleariae*, *Diabrotica* spp., *Psylliodes chrysocephala*, *Epilachna varivestis*, *Atomaria* spp., *Oryzaephilus surinamensis*, *Anthonomus* spp., *Sitophilus* spp., *Otiorrhynchus sulcatus*, *Cosmopolites sordidus*, *Ceuthorrhynchus assimilis*, *Hypera postica*, *Dermestes* spp., *Trogoderma* spp., *Anthrenus* spp., *Attagenus* spp., *Lyctus* spp., *Meligethes aeneus*, *Ptinus* spp., *Niptus hololeucus*, *Gibbium psyllodes*, *Tribolium* spp.,
30 *Tenebrio molitor*, *Agriotes* spp., *Conoderus* spp., *Melolontha melolontha*, *Amphimallon solstitialis*, *Costelytra zealandica* and *Lissorhoptrus oryzophilus*.

From the order of the Hymenoptera, for example, *Diprion* spp., *Hoplocampa* spp., *Lasius* spp., *Monomorium pharaonis* and *Vespa* spp.

- From the order of the Diptera, for example, *Aedes* spp., *Anopheles* spp., *Culex* spp.,
35 *Drosophila melanogaster*, *Musca* spp., *Fannia* spp., *Calliphora erythrocephala*, *Lucilia* spp., *Chrysomyia* spp., *Cuterebra* spp., *Gastrophilus* spp., *Hyppobosca* spp., *Stomoxys* spp., *Oestrus*

spp., Hypoderma spp., Tabanus spp., Tannia spp., Bibio hortulanus, Oscinella frit, Phorbia spp., Pegomyia hyoscyami, Ceratitis capitata, Dacus oleae, Tipula paludosa, Hylemyia spp. and Liriomyza spp.

From the order of the Siphonaptera, for example, Xenopsylla cheopis and Ceratophyllus spp.

5 From the class of the Arachnida, for example, Scorpio maurus, Latrodectus mactans, Acarus siro, Argas spp., Ornithodoros spp., Dermanyssus gallinae, Eriophyes ribis, Phyllocoptruta oleivora, Boophilus spp., Rhipicephalus spp., Amblyomma spp., Hyalomma spp., Ixodes spp., Psoroptes spp., Chorioptes spp., Sarcoptes spp., Tarsonemus spp., Bryobia praetiosa, Panonychus spp., Tetranychus spp., Hemitarsonemus spp., Brevipalpus spp.

10

The phytoparasitic nematodes include, for example, Pratylenchus spp., Radopholus similis, Ditylenchus dipsaci, Tylenchulus semipenetrans, Heterodera spp., Globodera spp., Meloidogyne spp., Aphelenchoides spp., Longidorus spp., Xiphinema spp., Trichodorus spp., Bursaphelenchus spp.

15

The compounds of the formula (I) according to the invention have in particular excellent activity against larvae of the mustard beetle (Phaedon cochleariae), against caterpillars of the diamond-back moth (Plutella xylostella), against caterpillars of the armyworm (Spodoptera frugiperda) and against caterpillars of the cotton bollworm (Heliothis armigera).

20

If appropriate, the compounds according to the invention can, at certain concentrations or application rates, also be used as herbicides or microbicides, for example as fungicides, antimycotics and bactericides. If appropriate, they can also be employed as intermediates or precursors for the synthesis of other active compounds.

25

All plants and plant parts can be treated in accordance with the invention. Plants are to be understood as meaning in the present context all plants and plant populations such as desired and undesired wild plants or crop plants (including naturally occurring crop plants). Crop plants can be plants which can be obtained by conventional plant breeding and optimization methods or by biotechnological and genetic engineering methods or by combinations of these methods, including the transgenic plants and including the plant cultivars protectable or not protectable by plant breeders' rights. Plant parts are to be understood as meaning all parts and organs of plants above and below the ground, such as shoot, leaf, flower and root, examples which may be mentioned being leaves, needles, stalks, stems, flowers, fruit bodies, fruits, seeds, roots, tubers and rhizomes. The plant parts also include harvested material, and

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vegetative and generative propagation material, for example cuttings, tubers, rhizomes, offshoots and seeds.

5 Treatment according to the invention of the plants and plant parts with the active compounds is carried out directly or by allowing the compounds to act on the surroundings, environment or storage space by the customary treatment methods, for example by immersion, spraying, evaporation, fogging, scattering, painting on, injection and, in the case of propagation material, in particular in the case of seeds, also by applying one or more coats.

10 The active compounds can be converted to the customary formulations, such as solutions, emulsions, wettable powders, suspensions, powders, dusts, pastes, soluble powders, granules, suspension-emulsion concentrates, natural and synthetic materials impregnated with active compound and microencapsulations in polymeric substances.

15 These formulations are produced in a known manner, for example by mixing the active compounds with extenders, that is liquid solvents, and/or solid carriers, optionally with the use of surfactants, that is emulsifiers and/or dispersants, and/or foam-formers.

20 If the extender used is water, it is also possible to employ for example organic solvents as auxiliary solvents. Essentially, suitable liquid solvents are: aromatics such as xylene, toluene or alkylnaphthalenes, chlorinated aromatics or chlorinated aliphatic hydrocarbons such as chlorobenzenes, chloroethylenes or methylene chloride, aliphatic hydrocarbons such as cyclohexane or paraffins, for example petroleum fractions, mineral and vegetable oils, alcohols
25 such as butanol or glycol and also their ethers and esters, ketones such as acetone, methyl ethyl ketone, methyl isobutyl ketone or cyclohexanone, strongly polar solvents such as dimethylformamide and dimethyl sulphoxide, and also water.

As solid carriers there are suitable:

30 for example ammonium salts and ground natural minerals such as kaolins, clays, talc, chalk, quartz, attapulgit, montmorillonite or diatomaceous earth, and ground synthetic minerals, such as highly disperse silica, alumina and silicates; as solid carriers for granules there are suitable: for example crushed and fractionated natural rocks such as calcite, marble, pumice, sepiolite and dolomite, and also synthetic granules of inorganic and organic meals, and granules of
35 organic material such as sawdust, coconut shells, maize cobs and tobacco stalks; as emulsifiers and/or foam-formers there are suitable: for example nonionic and anionic emulsifiers, such as

polyoxyethylene fatty acid esters, polyoxyethylene fatty alcohol ethers, for example alkylaryl polyglycol ethers, alkylsulphonates, alkyl sulphates, arylsulphonates and also protein hydrolysates; as dispersants there are suitable: for example lignosulphite waste liquors and methylcellulose.

5

Tackifiers such as carboxymethylcellulose and natural and synthetic polymers in the form of powders, granules or latices, such as gum arabic, polyvinyl alcohol and polyvinyl acetate, as well as natural phospholipids such as cephalins and lecithins, and synthetic phospholipids, can be used in the formulations. Other additives can be mineral and vegetable oils.

10

It is possible to use colorants such as inorganic pigments, for example iron oxide, titanium oxide and Prussian Blue, and organic dyestuffs, such as alizarin dyestuffs, azo dyestuffs and metal phthalocyanine dyestuffs, and trace nutrients such as salts of iron, manganese, boron, copper, cobalt, molybdenum and zinc.

15

The formulations generally comprise between 0.1 and 95% by weight of active compound, preferably between 0.5 and 90%.

20

The active compound according to the invention can be used in its commercially available formulations and in the use forms, prepared from these formulations, as a mixture with other active compounds, such as insecticides, attractants, sterilizing agents, bactericides, acaricides, nematocides, fungicides, growth-regulating substances or herbicides. The insecticides include, for example, phosphoric acid esters, carbamates, carboxylates, chlorinated hydrocarbons, phenylureas and substances produced by microorganisms, inter alia.

25

Particularly favourable mixing components are, for example, the following compounds:

Fungicides:

2-phenylphenol; 8-hydroxyquinoline sulphate; acibenzolar-S-methyl; aldimorph; amidoflumet; ampropylfos; ampropylfos-potassium; andoprim; anilazine; azaconazole; azoxystrobin; benalaxyl; benodanil; benomyl; benthiavalicarb-isopropyl; benzamacril; benzamacril-isobutyl; bilanafos; binapacryl; biphenyl; bitertanol; blasticidin-S; bromuconazole; bupirimate; buthiobate; butylamine; calcium polysulphide; capsimycin; captafol; captan; carbendazim; carboxin; carpropamid; carvone; chinomethionat; chlobenthiazone; chlorfenazole; chloroneb; chlorothalonil; chlozolate; clozylacon; cyazofamid; cyflufenamid; cymoxanil; cyproconazole; cyprodinil; cyprofuram; Dagger G; debacarb; dichlofluanid; dichlone; dichlorophen; diclocymet; diclomezine; dicloran; diethofencarb;

35

difenoconazole; diflumetorim; dimethirimol; dimethomorph; dimoxystrobin; diniconazole;
 diniconazole-M; dinocap; diphenylamine; dipyrithione; ditalimfos; dithianon; dodine;
 drazoxolon; edifenphos; epoxiconazole; ethaboxam; ethirimol; etridiazole; famoxadone;
 fenamidone; fenapanil; fenarimol; fenbuconazole; fenfuram; fenhexamid; fenitropan;
 5 fenoxanil; fenpiclonil; fenpropidin; fenpropimorph; ferbam; fluazinam; flubenzimine;
 fludioxonil; flumetover; flumorph; fluoromide; fluoxastrobin; fluquinconazole; flurprimidol;
 flusilazole; flusulfamide; flutolanil; flutriafol; folpet; fosetyl-Al; fosetyl-sodium;
 fuberidazole; furalaxyl; furametypr; furcarbanil; furmecyclox; guazatine;
 hexachlorobenzene; hexaconazole; hymexazole; imazalil; imibenconazole; iminoctadine
 10 triacetate; iminoctadine tris(albesilate); iodocarb; ipconazole; iprobenfos; iprodione;
 iprovalicarb; irumamycin; isoprothiolane; isovaledione; kasugamycin; kresoxim-methyl;
 mancozeb; maneb; meferimzone; mepanipyrim; mepronil; metalaxyl; metalaxyl-M; met-
 conazole; methasulfocarb; methfuroxam; metiram; metominostrobin; metsulfovax;
 mildiomyacin; myclobutanil; myclozolin; natamycin; nicobifen; nitrothal-isopropyl;
 15 noviflumuron; nuarimol; ofurace; orysastrobin; oxadixyl; oxolinic acid; oxpoconazole;
 oxycarboxin; oxyfenthiin; paclobutrazole; pefurazoate; penconazole; pencycuron;
 phosdiphen; phthalide; picoxystrobin; piperalin; polyoxins; polyoxorim; probenazole;
 prochloraz; procymidone; propamocarb; propanosine-sodium; propiconazole; propineb;
 proquinazid; prothioconazole; pyraclostrobin; pyrazophos; pyrifenox; pyrimethanil;
 20 pyroquilon; pyroxyfur; pyrrolnitrin; quinconazole; quinoxifen; quintozone; simeconazole;
 spiroxamine; sulphur; tebuconazole; tecloftalam; tecnazene; tetcyclacis; tetraconazole;
 thiabendazole; thicyofen; thifluzamide; thiophanate-methyl; thiram; tioxymid; tolclofos-
 methyl; tolylfluand; triadimefon; triadimenol; triazbutil; triazoxide; tricyclamide; tri-
 cyclazole; tridemorph; trifloxystrobin; triflumizole; triforine; triticonazole; uniconazole;
 25 validamycin A; vinclozolin; zineb; ziram; zoxamide; (2S)-N-[2-[4-[[3-(4-chlorophenyl)-2-
 propynyl]oxy]-3-methoxyphenyl]ethyl]-3-methyl-2-[(methylsulphonyl)amino]butanemide;
 1-(1-naphthalenyl)-1H-pyrrole-2,5-dione; 2,3,5,6-tetrachloro-4-(methylsulphonyl)pyridine;
 2-amino-4-methyl-N-phenyl-5-thiazolecarboxamide; 2-chloro-N-(2,3-dihydro-1,1,3-
 trimethyl-1H-inden-4-yl)-3-pyridinecarboxamide; 3,4,5-trichloro-2,6-pyridinedicarbonitrile;
 30 actinovate; cis-1-(4-chlorophenyl)-2-(1H-1,2,4-triazol-1-yl)cycloheptanol; methyl 1-(2,3-
 dihydro-2,2-dimethyl-1H-inden-1-yl)-1H-imidazole-5-carboxylate; monopotassium
 carbonate; N-(6-methoxy-3-pyridinyl)cyclopropanecarboxamide; N-butyl-8-(1,1-
 dimethylethyl)-1-oxaspiro[4.5]decane-3-amine; sodium tetrathiocarbonate;
 35 and copper salts and preparations, such as Bordeaux mixture; copper hydroxide; copper
 naphthenate; copper oxychloride; copper sulphate; cufraneb; copper oxide; mancopper;

oxine-copper.

Bactericides:

5 bronopol, dichlorophen, nitrapyrin, nickel dimethyldithiocarbamate, kasugamycin, othilinone, furancarboxylic acid, oxytetracyclin, probenazole, streptomycin, tecloftalam, copper sulphate and other copper preparations.

Insecticides / acaricides / nematocides:

10 abamectin, ABG-9008, acephate, acequinocyl, acetamiprid, acetoprole, acrinathrin, AKD-1022, AKD-3059, AKD-3088, alanycarb, aldicarb, aldoxycarb, allethrin, allethrin 1R-isomers, alpha-cypermethrin (alphamethrin), amidoflumet, aminocarb, amitraz, avermectin, AZ-60541, azadirachtin, azamethiphos, azinphos-methyl, azinphos-ethyl, azocyclotin, Bacillus popilliae, Bacillus sphaericus, Bacillus subtilis, Bacillus thuringiensis, Bacillus thuringiensis strain EG-2348, Bacillus thuringiensis strain GC-91, Bacillus thuringiensis strain NCTC-11821, baculoviruses, Beauveria bassiana, Beauveria tenella, bendiocarb, ben-
15 furacarb, bensultap, benzoximate, beta-cyfluthrin, beta-cypermethrin, bifenazate, bifenthrin, binapacryl, bioallethrin, bioallethrin-S-cyclopentyl-isomer, bioethanomethrin, biopermethrin, bioresmethrin, bistrifluron, BPMC, brofenprox, bromophos-ethyl, bromopropylate, bromfenvinfos (-methyl), BTG-504, BTG-505, bufencarb, buprofezin, butathiofos, butocarb-
20 oxim, butoxycarboxim, butylpyridaben, cadusafos, camphechlor, carbaryl, carbofuran, carbophenothion, carbosulfan, cartap, CGA-50439, chinomethionat, chlordane, chlordimeform, chloethocarb, chlorethoxyfos, chlorfenapyr, chlorfenvinphos, chlorfluazuron, chlor-mephos, chlorobenzilate, chloropicrin, chlorproxyfen, chlorpyrifos-methyl, chlorpyrifos (-ethyl), chlovaporthrin, chromafenozide, cis-cypermethrin, cis-resmethrin, cis-permethrin, clocythr-
25 in, cloethocarb, clofentezine, clothianidin, clothiazoben, codlemone, coumaphos, cyanofenphos, cyanophos, cycloprene, cycloprothrin, Cydia pomonella, cyfluthrin, cyhalothrin, cyhexatin, cypermethrin, cyphenothrin (1R-trans-isomer), cyromazine, DDT, delta-methrin, demeton-S-methyl, demeton-S-methylsulphone, diafenthiuron, dialifos, diazinon, dichlofenthion, dichlorvos, dicofol, dicrotophos, dicyclanil, diflubenzuron, dimethoate, di-
30 methylvinphos, dinobuton, dinocap, dinotefuran, diofenolan, disulfoton, docusat-sodium, dofenapyn, DOWCO-439, eflusilanate, emamectin, emamectin-benzoate, empen-
thrin (1R-isomer), endosulfan, Entomophthora spp., EPN, esfenvalerate, ethiofencarb, ethiprole, ethion, ethoprophos, etofenprox, etoxazole, etrimfos, famphur, fenamiphos, fenazaquin, fenbutatin oxide, fenfluthrin, fenitrothion, fenobucarb, fenothiocarb, fenoxacrim, fenoxycarb, fen-
35 propathrin, fenpyrad, fenpyrithrin, fenpyroximate, fensulfothion, fenthion, fentrifanil, fenvalerate, fipronil, flonicamid, fluacrypyrim, fluazuron, flubenzimine, flubrocylthrin, flu-

cycloxuron, flucythrinate, flufenimer, flufenoxuron, flufenprox, flumethrin, flupyrzofos, flutenzin (flufenzine), fluvalinate, fonofos, formetanate, formothion, fosmethilan, fos-thiazate, fubfenprox (fluproxyfen), furathiocarb, gamma-HCH, gossyplure, grandlure, granulosis viruses, halfenprox, halofenozide, HCH, HCN-801, heptenophos, hexaflumuron, 5 hexythiazox, hydramethylnone, hydroprene, IKA-2002, imidacloprid, imiprothrin, indoxa-carb, iodofenphos, iprobenfos, isazofos, isofenphos, isoprocarb, isoxathion, ivermectin, japonilure, kadethrin, nuclear polyhedrosis viruses, kinoprene, lambda-cyhalothrin, lindane, lufenuron, malathion, mecarbam, mesulfenfos, metaldehyde, metam-sodium, methacrifos, methamidophos, Metharhizium anisopliae, Metharhizium flavoviride, methidathion, methio- 10 carb, methomyl, methoprene, methoxychlor, methoxyfenozide, metolcarb, metoxadiazone, mevinphos, milbemectin, milbemycin, MKI-245, MON-45700, monocrotophos, moxidectin, MTI-800, naled, NC-104, NC-170, NC-184, NC-194, NC-196, niclosamide, nicotine, niten-pyram, nithiazine, NNI-0001, NNI-0101, NNI-0250, NNI-9768, novaluron, noviflumuron, OK-5101, OK-5201, OK-9601, OK-9602, OK-9701, OK-9802, omethoate, oxamyl, oxy- 15 demeton-methyl, Paecilomyces fumosoroseus, parathion-methyl, parathion (-ethyl), permethrin (cis-, trans-), petroleum, PH-6045, phenothrin (1R-trans isomer), phenthoate, phorate, phosalone, phosmet, phosphamidon, phosphocarb, phoxim, piperonyl butoxide, pirimicarb, pirimiphos-methyl, pirimiphos-ethyl, prallethrin, profenofos, promecarb, propa-phos, propargite, propetamphos, propoxur, prothiofos, prothoate, protrifenbute, pymetro- 20 zine, pyraclofos, pyresmethrin, pyrethrum, pyridaben, pyridalyl, pyridaphenthion, pyrida-thion, pyrimidifen, pyriproxyfen, quinalphos, resmethrin, RH-5849, ribavirin, RU-12457, RU-15525, S-421, S-1833, salithion, sebufos, SI-0009, silafluofen, spinosad, spiroadiclofen, spiromesifen, sulfluramid, sulfotep, sulprofos, SZI-121, tau-fluvalinate, tebufenozide, tebu-fenpyrad, tebupirimfos, teflubenzuron, tefluthrin, temephos, temiviphos, terbam, terbufos, 25 tetrachlorvinphos, tetradifon, tetramethrin, tetramethrin (1R-isomer), tetrasul, theta-cyper-methrin, thiacloprid, thiamethoxam, thiapronil, thiatriphos, thiocyclam hydrogenoxalate, thiodicarb, thiofanox, thiometon, thiosultap-sodium, thuringiensin, tolfenpyrad, tralocyttrin, tralomethrin, transfluthrin, triarathene, triazamate, triazophos, triazuron, trichlophenidine, trichlorfon, triflumuron, trimethacarb, vamidothion, vaniliprole, verbutin, Verticillium 30 lecanii, WL-108477, WL-40027, YI-5201, YI-5301, YI-5302, XMC, xylylcarb, ZA-3274, zeta-cypermethrin, zolaprofos, ZXI-8901, the compound 3-methylphenyl propylcarbamate (tsumacide Z), the compound 3-(5-chloro-3-pyridinyl)-8-(2,2,2-trifluoroethyl)-8-azabicyclo[3.2.1]octane-3-carbonitrile (CAS-Reg. No. 185982-80-3) and the corresponding 3-endo-isomer (CAS-Reg. No. 185984-60-5) (cf. WO-96/37494, WO-98/25923), and 35 preparations which comprise insecticidally active plant extracts, nematodes, fungi or viruses.

A mixture with other known active compounds, such as herbicides, fertilizers, growth regulators, safeners and/or semiochemicals is also possible.

5 When used as insecticides, the active compounds according to the invention can furthermore be present in their commercially available formulations and in the use forms, prepared from these formulations, as a mixture with synergistic agents. Synergistic agents are compounds, which increase the action of the active compounds, without it being necessary for the synergistic agent added to be active itself.

10 When used as insecticides, the active compounds according to the invention can furthermore be present in their commercially available formulations and in the use forms, prepared from these formulations, as a mixture with inhibitors which reduce degradation of the active compound after use in the vicinity of the plant, on the surface of parts of plants or in plant
15 tissues.

The active compound content of the use forms prepared from the commercially available formulations can vary within wide limits. The active compound concentration of the use forms can be from 0.0000001 to 95% by weight of active compound, preferably between
20 0.0001 and 1% by weight.

The compounds are employed in a customary manner appropriate for the use forms.

25 When used against hygiene pests and pests of stored products, the active compound is distinguished by an excellent residual action on wood and clay as well as a good stability to alkali on limed substrates.

30 As already mentioned above, it is possible to treat all plants and their parts according to the invention. In a preferred embodiment, wild plant species and plant cultivars, or those obtained by conventional biological breeding methods, such as crossing or protoplast fusion, and parts thereof, are treated. In a further preferred embodiment, transgenic plants and plant cultivars obtained by genetic engineering, if appropriate in combination with conventional methods (Genetically Modified Organisms), and parts thereof are treated. The term "parts" or "parts of plants" or "plant parts" has been explained above.

Particularly preferably, plants of the plant cultivars which are in each case commercially available or in use are treated according to the invention. Plant cultivars are to be understood as meaning plants having novel properties ("traits") which have been obtained by conventional breeding, by mutagenesis or by recombinant DNA techniques. These can be cultivars, bio- or genotypes.

Depending on the plant species or plant cultivars, their location and growth conditions (soils, climate, vegetation period, diet), the treatment according to the invention may also result in superadditive ("synergistic") effects. Thus, for example, reduced application rates and/or a widening of the activity spectrum and/or an increase in the activity of the substances and compositions to be used according to the invention, better plant growth, increased tolerance to high or low temperatures, increased tolerance to drought or to water or soil salt content, increased flowering performance, easier harvesting, accelerated maturation, higher harvest yields, better quality and/or a higher nutritional value of the harvested products, better storage stability and/or processability of the harvested products are possible which exceed the effects which were actually to be expected.

The transgenic plants or plant cultivars (i.e. those obtained by genetic engineering) which are preferably to be treated according to the invention include all plants which, in the genetic modification, received genetic material which imparted particularly advantageous, useful traits to these plants. Examples of such traits are better plant growth, increased tolerance to high or low temperatures, increased tolerance to drought or to water or soil salt content, increased flowering performance, easier harvesting, accelerated maturation, higher harvest yields, better quality and/or a higher nutritional value of the harvested products, better storage stability and/or processability of the harvested products. Further and particularly emphasized examples of such traits are a better defence of the plants against animal and microbial pests, such as against insects, mites, phytopathogenic fungi, bacteria and/or viruses, and also increased tolerance of the plants to certain herbicidally active compounds. Examples of transgenic plants which may be mentioned are the important crop plants, such as cereals (wheat, rice), maize, soya beans, potatoes, cotton, tobacco, oilseed rape and also fruit plants (with the fruits apples, pears, citrus fruits and grapes), and particular emphasis is given to maize, soya beans, potatoes, cotton, tobacco and oilseed rape. Traits that are emphasized are in particular increased defence of the plants against insects, arachnids, nematodes and worms by toxins formed in the plants, in particular those formed in the plants by the genetic material from *Bacillus thuringiensis* (for example by the genes CryIA(a), CryIA(b), CryIA(c), CryIIA, CryIIIA, CryIIIB2, Cry9c, Cry2Ab, Cry3Bb and

CryIF and also combinations thereof) (hereinbelow referred to as "Bt plants"). Traits that are also particularly emphasized are the increased defence of the plants against fungi, bacteria and viruses by systemic acquired resistance (SAR), systemin, phytoalexins, elicitors and resistance genes and correspondingly expressed proteins and toxins. Traits that are

5 furthermore particularly emphasized are the increased tolerance of the plants to certain herbicidally active compounds, for example imidazolinones, sulphonylureas, glyphosate or phosphinotricin (for example the "PAT" gene). The genes which impart the desired traits in question can also be present in combination with one another in the transgenic plants. Examples of "Bt plants" which may be mentioned are maize varieties, cotton varieties, soya

10 bean varieties and potato varieties which are sold under the trade names YIELD GARD® (for example maize, cotton, soya beans), KnockOut® (for example maize), StarLink® (for example maize), Bollgard® (cotton), Nucotn® (cotton) and NewLeaf® (potato). Examples of herbicide-tolerant plants which may be mentioned are maize varieties, cotton varieties and soya bean varieties which are sold under the trade names Roundup Ready® (tolerance

15 to glyphosate, for example maize, cotton, soya bean), Liberty Link® (tolerance to phosphinotricin, for example oilseed rape), IMI® (tolerance to imidazolinones) and STS® (tolerance to sulphonylureas, for example maize). Herbicide-resistant plants (plants bred in a conventional manner for herbicide tolerance) which may be mentioned include the varieties sold under the name Clearfield® (for example maize). Of course, these statements also

20 apply to plant cultivars having these genetic traits or genetic traits still to be developed, which plants will be developed and/or marketed in the future.

The plants listed can be treated according to the invention in a particularly advantageous manner with the compounds of the general formula (I) and/or the active compound mixtures

25 according to the invention. The preferred ranges stated above for the active compounds or mixtures also apply to the treatment of these plants. Particular emphasis is given to the treatment of plants with the compounds or mixtures specifically mentioned in the present text.

30 The active compounds according to the invention act not only against plant, hygiene and stored product pests, but also in the veterinary medicine sector against animal parasites (ectoparasites), such as hard ticks, soft ticks, mange mites, leaf mites, flies (biting and licking), parasitic fly larvae, lice, hair lice, feather lice and fleas. These parasites include:

35 From the order of the Anoplurida, for example, Haematopinus spp., Linognathus spp., Pediculus spp., Phtirus spp. and Solenopotes spp.

From the order of the Mallophagida and the suborders Amblycerina and Ischnocerina, for example, Trimenopon spp., Menopon spp., Trinoton spp., Bovicola spp., Werneckiella spp., Lepikentron spp., Damalina spp., Trichodectes spp. and Felicola spp.

5 From the order of the Diptera and the suborders Nematocerina and Brachycerina, for example, Aedes spp., Anopheles spp., Culex spp., Simulium spp., Eusimulium spp., Phlebotomus spp., Lutzomyia spp., Culicoides spp., Chrysops spp., Hybomitra spp., Atylotus spp., Tabanus spp., Haematopota spp., Philipomyia spp., Braula spp., Musca spp., Hydrotaea spp., Stomoxys spp., Haematobia spp., Morellia spp., Fannia spp., Glossina spp., Calliphora spp., Lucilia spp., Chrysomyia spp., Wohlfahrtia spp., Sarcophaga spp., Oestrus
10 spp., Hypoderma spp., Gasterophilus spp., Hippobosca spp., Lipoptena spp. and Melophagus spp.

From the order of the Siphonaptera, for example Pulex spp., Ctenocephalides spp., Xenopsylla spp. and Ceratophyllus spp.

15 From the order of the Heteroptera, for example, Cimex spp., Triatoma spp., Rhodnius spp. and Panstrongylus spp.

From the order of the Blattellidae, for example Blatta orientalis, Periplaneta americana, Blattella germanica and Supella spp.

20 From the subclass of the Acari (Acarida) and the orders of the Meta- and Mesostigmata, for example, Argas spp., Ornithodoros spp., Otobius spp., Ixodes spp., Amblyomma spp., Boophilus spp., Dermacentor spp., Haemophysalis spp., Hyalomma spp., Rhipicephalus spp., Dermanyssus spp., Raillietia spp., Pneumonyssus spp., Sternostoma spp. and Varroa spp.

25 From the order of the Actiniedida (Prostigmata) und Acaridida (Astigmata), for example, Acarapis spp., Cheyletiella spp., Ornithocheyletiella spp., Myobia spp., Psorergates spp., Demodex spp., Trombicula spp., Listrophorus spp., Acarus spp., Tyrophagus spp., Caloglyphus spp., Hypodectes spp., Pterolichus spp., Psoroptes spp., Chorioptes spp., Otodectes spp., Sarcoptes spp., Notoedres spp., Knemidocoptes spp., Cytodites spp. and Laminosioptes spp.

30 The active compounds of the formula (I) according to the invention are also suitable for controlling arthropods which infest agricultural productive livestock, such as, for example, cattle, sheep, goats, horses, pigs, donkeys, camels, buffalo, rabbits, chickens, turkeys, ducks, geese and bees, other pets, such as, for example, dogs, cats, caged birds and aquarium fish, and also so-called test animals, such as, for example, hamsters, guinea pigs, rats and mice.

35 By controlling these arthropods, cases of death and reduction in productivity (for meat, milk,

wool, hides, eggs, honey etc.) should be diminished, so that more economic and easier animal husbandry is possible by use of the active compounds according to the invention.

5 The active compounds according to the invention are used in the veterinary sector in a known manner by enteral administration in the form of, for example, tablets, capsules, potions, drenches, granules, pastes, boluses, the feed-through process and suppositories, by parenteral administration, such as, for example, by injection (intramuscular, subcutaneous, intravenous, intraperitoneal and the like), implants by nasal administration, by dermal use in the form, for example, of dipping or bathing, spraying, pouring on and spotting on, washing
10 and powdering, and also with the aid of moulded articles containing the active compound, such as collars, ear marks, tail marks, limb bands, halters, marking devices and the like.

When used for cattle, poultry, pets and the like, the active compounds of the formula (I) can be used as formulations (for example powders, emulsions, free-flowing compositions),
15 which comprise the active compounds in an amount of 1 to 80% by weight, directly or after 100 to 10 000-fold dilution, or they can be used as a chemical bath.

It has furthermore been found that the compounds according to the invention also have a strong insecticidal action against insects which destroy industrial materials.
20

The following insects may be mentioned as examples and as preferred - but without a limitation:

25 Beetles, such as *Hylotrupes bajulus*, *Chlorophorus pilosis*, *Anobium punctatum*, *Xestobium rufovillosum*, *Ptilinus pecticornis*, *Dendrobium pertinex*, *Ernobius mollis*, *Priobium carpini*, *Lyctus brunneus*, *Lyctus africanus*, *Lyctus planicollis*, *Lyctus linearis*, *Lyctus pubescens*, *Trogoxylon aequale*, *Minthes rugicollis*, *Xyleborus spec.*, *Tryptodendron spec.*, *Apate monachus*, *Bostrychus capucins*, *Heterobostrychus brunneus*, *Sinoxylon spec.*, *Dinoderus minutus*;

30 Hymenopterons, such as *Sirex juvencus*, *Urocerus gigas*, *Urocerus gigas taignus*, *Urocerus augur*;

Termites, such as *Kaloterms flavicollis*, *Cryptoterms brevis*, *Heteroterms indicola*, *Reticuliterms flavipes*, *Reticuliterms santonensis*, *Reticuliterms lucifugus*, *Mastoterms darwiniensis*, *Zootermopsis nevadensis*, *Coptoterms formosanus*;

35 Bristletails, such as *Lepisma saccharina*.

Industrial materials in the present connection are to be understood as meaning non-living materials, such as, preferably, plastics, adhesives, sizes, papers and cards, leather, wood and processed wood products and coating compositions.

- 5 Wood and processed wood products are materials to be protected, especially preferably, from insect infestation.

Wood and processed wood products which can be protected by the agents according to the invention or mixtures comprising these are to be understood as meaning, for example:

10

building timber, wooden beams, railway sleepers, bridge components, boat jetties, wooden vehicles, boxes, pallets, containers, telegraph poles, wood panelling, wooden windows and doors, plywood, chipboard, joinery or wooden products which are used quite generally in house-building or in building joinery.

15

The active compounds can be used as such, in the form of concentrates or in generally customary formulations, such as powders, granules, solutions, suspensions, emulsions or pastes.

20

The formulations mentioned can be prepared in a manner known per se, for example by mixing the active compounds with at least one solvent or diluent, emulsifier, dispersing agent and/or binder or fixing agent, a water repellent, if appropriate siccatives and UV stabilizers and if appropriate dyestuffs and pigments, and also other processing auxiliaries.

25

The insecticidal compositions or concentrates used for the preservation of wood and wood-derived timber products comprise the active compound according to the invention in a concentration of 0.0001 to 95% by weight, in particular 0.001 to 60% by weight.

30

The amount of the compositions or concentrates employed depends on the nature and occurrence of the insects and on the medium. The optimum amount employed can be determined for the use in each case by a series of tests. In general, however, it is sufficient to employ 0.0001 to 20% by weight, preferably 0.001 to 10% by weight, of the active compound, based on the material to be preserved.

35

Solvents and/or diluents which are used are an organic chemical solvent or solvent mixture and/or an oily or oil-like organic chemical solvent or solvent mixture of low volatility and/or

a polar organic chemical solvent or solvent mixture and/or water, and if appropriate an emulsifier and/or wetting agent.

5 Organic chemical solvents which are preferably used are oily or oil-like solvents having an evaporation number above 35 and a flashpoint above 30°C, preferably above 45°C. Substances which are used as such oily or oil-like water-insoluble solvents of low volatility are appropriate mineral oils or aromatic fractions thereof, or solvent mixtures containing mineral oils, preferably white spirit, petroleum and/or alkylbenzene.

10 Mineral oils having a boiling range from 170 to 220°C, white spirit having a boiling range from 170 to 220°C, spindle oil having a boiling range from 250 to 350°C, petroleum and aromatics having a boiling range from 160 to 280°C, turpentine oil and the like, are advantageously employed.

15 In a preferred embodiment, liquid aliphatic hydrocarbons having a boiling range from 180 to 210°C or high-boiling mixtures of aromatic and aliphatic hydrocarbons having a boiling range from 180 to 220°C and/or spindle oil and/or monochloronaphthalene, preferably α -monochloronaphthalene, are used.

20 The organic oily or oil-like solvents of low volatility which have an evaporation number above 35 and a flashpoint above 30°C, preferably above 45°C, can be replaced in part by organic chemical solvents of high or medium volatility, provided that the solvent mixture likewise has an evaporation number above 35 and a flashpoint above 30°C, preferably above 45°C, and that the insecticide/fungicide mixture is soluble or emulsifiable in this solvent
25 mixture.

According to a preferred embodiment, some of the organic chemical solvent or solvent mixture is replaced by an aliphatic polar organic chemical solvent or solvent mixture. Aliphatic organic chemical solvents containing hydroxyl and/or ester and/or ether groups,
30 such as, for example, glycol ethers, esters or the like, are preferably used.

Organic chemical binders which are used in the context of the present invention are the synthetic resins and/or binding drying oils which are known per se, are water-dilutable and/or are soluble or dispersible or emulsifiable in the organic chemical solvents employed,
35 in particular binders consisting of or comprising an acrylate resin, a vinyl resin, for example polyvinyl acetate, polyester resin, polycondensation or polyaddition resin, polyurethane

resin, alkyd resin or modified alkyd resin, phenolic resin, hydrocarbon resin, such as indene-cumarone resin, silicone resin, drying vegetable oils and/or drying oils and/or physically drying binders based on a natural and/or synthetic resin.

5 The synthetic resin used as the binder can be employed in the form of an emulsion, dispersion or solution. Bitumen or bituminous substances can also be used as binders in an amount of up to 10% by weight. Dyestuffs, pigments, water-repelling agents, odour correctants and inhibitors or anticorrosive agents and the like which are known per se can additionally be employed.

10

It is preferred according to the invention for the composition or concentrate to comprise, as the organic chemical binder, at least one alkyd resin or modified alkyd resin and/or a drying vegetable oil. Alkyd resins having an oil content of more than 45% by weight, preferably 50 to 68% by weight, are preferably used according to the invention.

15

All or some of the binder mentioned can be replaced by a fixing agent (mixture) or a plasticizer (mixture). These additives are intended to prevent evaporation of the active compounds and crystallization or precipitation. They preferably replace 0.01 to 30% of the binder (based on 100% of the binder employed).

20

The plasticizers originate from the chemical classes of phthalic acid esters, such as dibutyl, dioctyl or benzyl butyl phthalate, phosphoric acid esters, such as tributyl phosphate, adipic acid esters, such as di-(2-ethylhexyl) adipate, stearates, such as butyl stearate or amyl stearate, oleates, such as butyl oleate, glycerol ethers or higher molecular weight glycol ethers, glycerol esters and p-toluenesulphonic acid esters.

25

Fixing agents are based chemically on polyvinyl alkyl ethers, such as, for example, polyvinyl methyl ether or ketones, such as benzophenone or ethylenebenzophenone.

30

Possible solvents or diluents are, in particular, also water, if appropriate as a mixture with one or more of the abovementioned organic chemical solvents or diluents, emulsifiers and dispersing agents.

35

Particularly effective preservation of wood is achieved by impregnation processes on a large industrial scale, for example vacuum, double vacuum or pressure processes.

The ready-to-use compositions can also comprise other insecticides, if appropriate, and also one or more fungicides, if appropriate.

Possible additional mixing partners are, preferably, the insecticides and fungicides mentioned in WO 94/29 268. The compounds mentioned in this document are an explicit constituent of the present application.

Especially preferred mixing partners which may be mentioned are insecticides, such as chlorpyrifos, phoxim, silafluofin, alphasmethrin, cyfluthrin, cypermethrin, deltamethrin, permethrin, imidacloprid, NI-25, flufenoxuron, hexaflumuron, transfluthrin, thiacloprid, methoxyfenozide, triflumuron, clothianidin, spinosad, tefluthrin, and also fungicides, such as epoxyconazole, hexaconazole, azaconazole, propiconazole, tebuconazole, cyproconazole, metconazole, imazalil, dichlorfluanid, tolylfluanid, 3-iodo-2-propinyl-butyl carbamate, N-octyl-isothiazolin-3-one and 4,5-dichloro-N-octylisothiazolin-3-one.

The compounds according to the invention can at the same time be employed for protecting objects which come into contact with saltwater or brackish water, such as hulls, screens, nets, buildings, moorings and signalling systems, against fouling.

Fouling by sessile Oligochaeta, such as Serpulidae, and by shells and species from the Ledamorpha group (goose barnacles), such as various Lepas and Scalpellum species, or by species from the Balanomorpha group (acorn barnacles), such as Balanus or Pollicipes species, increases the frictional drag of ships and, as a consequence, leads to a marked increase in operational costs owing to higher energy consumption and additionally frequent residence in the dry dock.

Apart from fouling by algae, for example Ectocarpus sp. and Ceramium sp., fouling by sessile Entomostraka groups, which come under the generic term Cirripedia (cirriped crustaceans), is of particular importance.

Surprisingly, it has now been found that the compounds according to the invention, alone or in combination with other active compounds, have an outstanding antifouling action.

Using the compounds according to the invention, alone or in combination with other active compounds, allows the use of heavy metals such as, for example, in bis(trialkyltin) sulphides, tri-*n*-butyltin laurate, tri-*n*-butyltin chloride, copper(I) oxide, triethyltin chloride,

tri-*n*-butyl(2-phenyl-4-chlorophenoxy)tin, tributyltin oxide, molybdenum disulphide, antimony oxide, polymeric butyl titanate, phenyl-(bispyridine)-bismuth chloride, tri-*n*-butyltin fluoride, manganese ethylenebisthiocarbamate, zinc dimethyldithiocarbamate, zinc ethylenebisthiocarbamate, zinc salts and copper salts of 2-pyridinethiol 1-oxide, bisdimethyldithiocarbamoylzinc ethylene-bisthiocarbamate, zinc oxide, copper(I) ethylene-bisdithiocarbamate, copper thiocyanate, copper naphthenate and tributyltin halides to be dispensed with, or the concentration of these compounds to be substantially reduced.

If appropriate, the ready-to-use antifouling paints can additionally comprise other active compounds, preferably algicides, fungicides, herbicides, molluscicides, or other antifouling active compounds.

Preferably suitable components in combinations with the antifouling compositions according to the invention are:

algicides such as 2-*tert*-butylamino-4-cyclopropylamino-6-methylthio-1,3,5-triazine, dichlorophen, diuron, endothal, fentin acetate, isoproturon, methabenzthiazuron, oxyfluorfen, quinoclamine and terbutryn;

fungicides such as benzo[*b*]thiophenecarboxylic acid cyclohexylamide S,S-dioxide, dichlofluanid, fluorfolpet, 3-iodo-2-propinyl butylcarbamate, tolylfluanid and azoles such as azaconazole, cyproconazole, epoxyconazole, hexaconazole, metconazole, propiconazole and tebuconazole;

molluscicides such as fentin acetate, metaldehyde, methiocarb, niclosamid, thiodicarb and trimethacarb, Fe-chelate;

or conventional antifouling active compounds such as 4,5-dichloro-2-octyl-4-isothiazolin-3-one, diiodomethylparatryl sulphone, 2-(N,N-dimethylthiocarbamoylthio)-5-nitrothiazyl, potassium, copper, sodium and zinc salts of 2-pyridinethiol 1-oxide, pyridine-triphenylborane, tetrabutyl-distannoxane, 2,3,5,6-tetrachloro-4-(methylsulphonyl)-pyridine, 2,4,5,6-tetrachloroisophthalonitrile, tetramethylthiuram disulphide and 2,4,6-trichlorophenylmaleimide.

The antifouling compositions used comprise the active compound according to the invention of the compositions according to the invention in a concentration of 0.001 to 50% by weight, in particular 0.01 to 20% by weight.

5 Moreover, the antifouling compositions according to the invention comprise the customary components such as, for example, those described in Ungerer, *Chem. Ind.* 1985, 37, 730-732 and Williams, *Antifouling Marine Coatings*, Noyes, Park Ridge, 1973.

Besides the algicidal, fungicidal, molluscicidal active compounds and insecticidal active
10 compounds according to the invention, antifouling paints comprise, in particular, binders.

Examples of recognized binders are polyvinyl chloride in a solvent system, chlorinated rubber in a solvent system, acrylic resins in a solvent system, in particular in an aqueous system, vinyl chloride/vinyl acetate copolymer systems in the form of aqueous dispersions
15 or in the form of organic solvent systems, butadiene/styrene/acrylonitrile rubbers, drying oils such as linseed oil, resin esters or modified hardened resins in combination with tar or bitumens, asphalt and epoxy compounds, small amounts of chlorine rubber, chlorinated polypropylene and vinyl resins.

20 If appropriate, paints also comprise inorganic pigments, organic pigments or colorants which are preferably insoluble in salt water. Paints may furthermore comprise materials such as colophonium to allow controlled release of the active compounds. Furthermore, the paints may comprise plasticizers, modifiers which affect the rheological properties and other conventional constituents. The compounds according to the invention or the
25 abovementioned mixtures may also be incorporated into self-polishing antifouling systems.

The active compounds are also suitable for controlling animal pests, in particular insects, arachnids and mites, which are found in enclosed spaces such as, for example, dwellings, factory halls, offices, vehicle cabins and the like. They can be employed alone or in
30 combination with other active compounds and auxiliaries in domestic insecticide products for controlling these pests. They are active against sensitive and resistant species and against all developmental stages. These pests include:

From the order of the Scorpionidea, for example, *Buthus occitanus*.

35 From the order of the Acarina, for example, *Argas persicus*, *Argas reflexus*, *Bryobia* spp., *Dermanyssus gallinae*, *Glyciphagus domesticus*, *Ornithodoros moubat*, *Rhipicephalus*

sanguineus, Trombicula alfreddugesi, Neutrombicula autumnalis, Dermatophagoides pteronissimus, Dermatophagoides forinae.

From the order of the Araneae, for example, Aviculariidae, Araneidae.

From the order of the Opiliones, for example, Pseudoscorpiones chelifer, Pseudoscorpiones cheiridium, Opiliones phalangium.

From the order of the Isopoda, for example, Oniscus asellus, Porcellio scaber.

From the order of the Diplopoda, for example, Blaniulus guttulatus, Polydesmus spp.

From the order of the Chilopoda, for example, Geophilus spp.

From the order of the Zygentoma, for example, Ctenolepisma spp., Lepisma saccharina, Lepismodes inquilinus.

From the order of the Blattaria, for example, Blatta orientalis, Blattella germanica, Blattella asahinai, Leucophaea maderae, Panchlora spp., Parcoblatta spp., Periplaneta australasiae, Periplaneta americana, Periplaneta brunnea, Periplaneta fuliginosa, Supella longipalpa.

From the order of the Saltatoria, for example, Acheta domesticus.

From the order of the Dermaptera, for example, Forficula auricularia.

From the order of the Isoptera, for example, Kaloterms spp., Reticulitermes spp.

From the order of the Psocoptera, for example, Lepinatus spp., Liposcelis spp.

From the order of the Coleoptera, for example, Anthrenus spp., Attagenus spp., Dermestes spp., Latheticus oryzae, Necrobia spp., Ptinus spp., Rhizopertha dominica, Sitophilus granarius, Sitophilus oryzae, Sitophilus zeamais, Stegobium paniceum.

From the order of the Diptera, for example, Aedes aegypti, Aedes albopictus, Aedes taeniorhynchus, Anopheles spp., Calliphora erythrocephala, Chrysosoma pluvialis, Culex quinquefasciatus, Culex pipiens, Culex tarsalis, Drosophila spp., Fannia canicularis, Musca domestica, Phlebotomus spp., Sarcophaga carnaria, Simulium spp., Stomoxys calcitrans, Tipula paludosa.

From the order of the Lepidoptera, for example, Achroia grisella, Galleria mellonella, Plodia interpunctella, Tinea cloacella, Tinea pellionella, Tineola bisselliella.

From the order of the Siphonaptera, for example, Ctenocephalides canis, Ctenocephalides felis, Pulex irritans, Tunga penetrans, Xenopsylla cheopis.

From the order of the Hymenoptera, for example, Camponotus herculeanus, Lasius fuliginosus, Lasius niger, Lasius umbratus, Monomorium pharaonis, Paravespula spp., Tetramorium caespitum.

From the order of the Anoplura, for example, Pediculus humanus capitis, Pediculus humanus corporis, Phthirus pubis.

From the order of the Heteroptera, for example, Cimex hemipterus, Cimex lectularius, Rhodinus prolixus, Triatoma infestans.

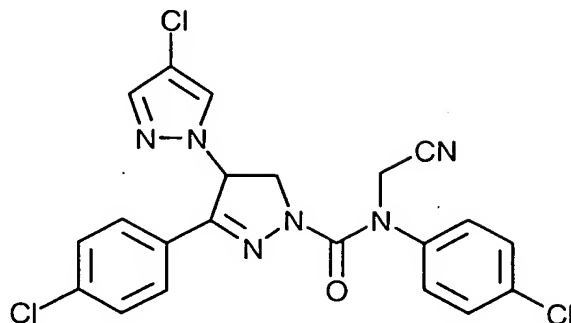
In the field of household insecticides, they are used alone or in combination with other suitable active compounds, such as phosphoric acid esters, carbamates, pyrethroids, neonicotinoides growth regulators or active compounds from other known classes of insecticides.

5

They are used as aerosols, pressure-free spray products, for example pump and atomizer sprays, automatic fogging systems, foggers, foams, gels, evaporator products with evaporator tablets made of cellulose or polymer, liquid evaporators, gel and membrane evaporators, propeller-driven evaporators, energy-free, or passive, evaporation systems, moth papers, moth bags and moth gels, as granules or dusts, in baits for spreading or in bait stations.

10

The preparation and use of the substances according to the invention are illustrated by the examples below.

Preparation Examples:**Example 1:**

5

Process (a)

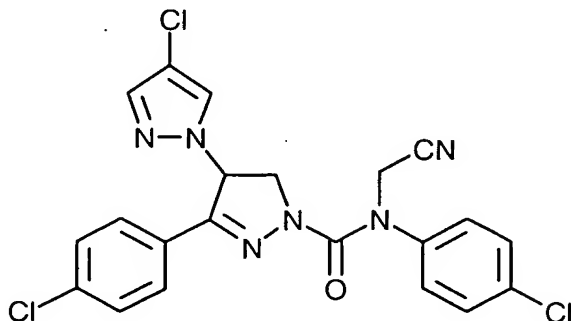
At 0°C, a solution of 26.6 g (0.16 mol) of N-cyanomethyl-4-chloroaniline and 16.5 g (0.16 mol) of triethylamine in 75 ml of methylene chloride is added dropwise to a solution of 16 g (54 mmol) of triphosgene (bistrichloromethyl carbamate) in 100 ml of methylene chloride, and the mixture is stirred at room temperature (about 20°C) for half an hour. A solution of 45.0 g (0.16 mol) of 3-(4-chlorophenyl)-4-(4-chloropyrazol-1-yl)-4,5-dihydro-1H-pyrazole and 16.5 g (0.16 mol) of triethylamine in 75 ml of methylene chloride is then added dropwise. The reaction mixture is stirred at room temperature for 18 hours and then washed twice with in each case 100 ml of water. The organic phase is dried over sodium sulphate and concentrated under reduced pressure. The residue is triturated with ethanol, filtered off with suction and washed with ethanol.

10

15

This gives 51.5 g (68% of theory) of 3-(4-chlorophenyl)-4-(4-chloropyrazol-1-yl)-4,5-dihydro-1-pyrazolecarboxylic acid (N-cyanomethyl)-4-chloroanilide of logP (pH 2.3) = 3.94.

20

Example 2:

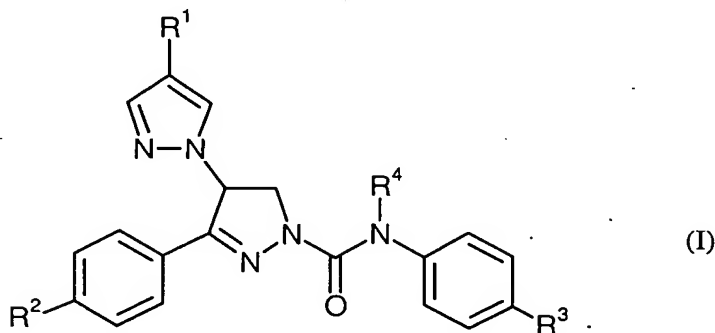
Process (b)

At 0°C, 2.78 g (10 mmol) of N-cyanomethyl-N-(4-chlorophenyl)carbonyl chloride are added to a mixture of 2.81 g (10 mmol) of 3-(4-chlorophenyl)-4-(4-chloropyrazol-1-yl)-4,5-dihydro-1H-pyrazole, 1.5 ml of triethylamine and 50 ml of methylene chloride. The mixture is stirred at room temperature (about 20°C) for 18 hours and then washed twice with in each case 30 ml of water. The organic phase is dried over sodium sulphate and concentrated under reduced pressure. The residue is triturated with ethanol, filtered off with suction and washed with ethanol.

This gives 3.7 g (79% of theory) of 3-(4-chlorophenyl)-4-(4-chloropyrazol-1-yl)-4,5-dihydro-1-pyrazolecarboxylic acid (N-cyanomethyl)-4-chloroanilide with logP (pH 2.3) = 3.94.

The compounds of the formula (I) listed in table 1 below can also be obtained analogously to examples 1 and 2 and/or in accordance with the general statements on the preparation:

Table 1:

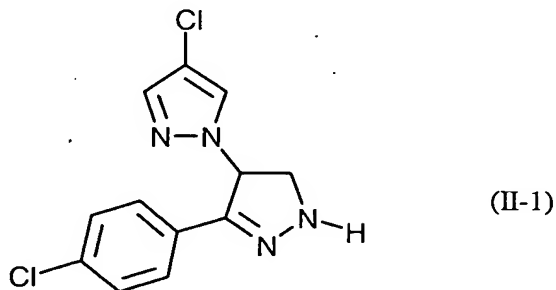


Ex. No.	R ¹	R ²	R ³	R ⁴
3	Cl	Cl	CN	CH ₂ CN
4	Cl	Cl	Br	CH ₂ CN
5	Cl	Cl	SCF ₃	CH ₂ CN
6	Cl	Cl	OCF ₃	CH ₂ CN
7	Cl	Cl	CF ₃	CH ₂ CN
8	Cl	Br	Cl	CH ₂ CN
9	Cl	Br	CN	CH ₂ CN
10	Cl	Br	Br	CH ₂ CN
11	Cl	Br	SCF ₃	CH ₂ CN
12	Cl	Br	OCF ₃	CH ₂ CN
13	Cl	Br	CF ₃	CH ₂ CN

Ex. No.	R ¹	R ²	R ³	R ⁴
14	Cl	F	Cl	CH ₂ CN
15	Cl	F	CN	CH ₂ CN
16	Cl	F	Br	CH ₂ CN
17	Cl	F	SCF ₃	CH ₂ CN
18	Cl	F	OCF ₃	CH ₂ CN
19	Cl	F	CF ₃	CH ₂ CN
20	Br	Cl	CN	CH ₂ CN
21	Br	Cl	Br	CH ₂ CN
22	Br	Cl	SCF ₃	CH ₂ CN
23	Br	Cl	OCF ₃	CH ₂ CN
24	Br	Cl	CF ₃	CH ₂ CN
25	Br	Br	Cl	CH ₂ CN
26	Br	Br	CN	CH ₂ CN
27	Br	Br	Br	CH ₂ CN
28	Br	Br	SCF ₃	CH ₂ CN
29	Br	Br	OCF ₃	CH ₂ CN
30	Br	Br	CF ₃	CH ₂ CN
31	Br	F	Cl	CH ₂ CN
32	Br	F	CN	CH ₂ CN
33	Br	F	Br	CH ₂ CN
34	Br	F	SCF ₃	CH ₂ CN
35	Br	F	OCF ₃	CH ₂ CN
36	Br	F	CF ₃	CH ₂ CN
37	Br	Cl	Cl	CH ₂ CN

Preparation of the starting materials of formula (II)

Example (II-1):



5

At room temperature (about 20°C), 2.16 g (0.021 mol) of bisdimethylaminomethane are added to a solution of 5.1 g (0.02 mol) of 2-(4-chloropyrazol-1-yl)-4'-chloroacetophenone in 50 ml of methylene chloride, and the mixture is heated under reflux for 18 hours. The

solvent is then distilled off under reduced pressure and the residue is dissolved in 50 ml of ethanol. Following the addition of 1.13 g (0.0226 mol) of hydrazine hydrate, the reaction mixture is stirred at 30°C for 3 hours. The precipitated product is filtered off with suction and washed with a little cold ethanol and with water.

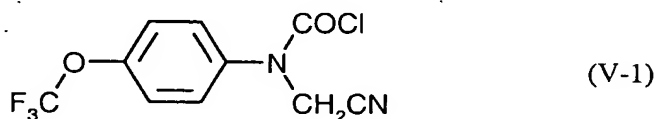
5

This gives 3.42 g (61% of theory) of 3-(4-chlorophenyl)-4-(4-chloropyrazol-1-yl)-4,5-dihydro-1H-pyrazole of logP (pH 2.3) = 2.64.

Preparation of the starting materials of the formula (V)

10

Example (V-1)



15

At 0°C, a solution of 20.7 g (0.0958 mol) of N-cyanomethyl-4-trifluoromethoxyaniline and 11.6 g (0.115 mol) of triethylamine in 150 ml of toluene is added dropwise to a solution of 10.4 g (0.105 mol) of phosgene in 100 ml of toluene, and the mixture is stirred at room temperature (about 20°C) for 18 hours. Excess phosgene is then flushed out. The mixture is washed with water, the organic phase is dried over sodium sulphate and the solvent is then distilled off under reduced pressure.

20

This gives 26.3 g (94% of theory) of N-cyanomethyl-N-(4-trifluoromethoxy)phenyl-carbamoyl chloride having the refractive index $n_D^{20} = 1.4816$.

25

The logP values given in the Preparation Examples are determined in accordance with EEC Directive 79/831 Annex V.A8 by HPLC (High Performance Liquid Chromatography) using a reversed-phase column (C 18). Temperature: 43°C.

30

The determination is carried out in the acidic range at pH 2.3 using the mobile phases 0.1% aqueous phosphoric acid and acetonitrile; linear gradient from 10% acetonitrile to 90% acetonitrile.

Calibration is carried out using unbranched alkan-2-ones (having 3 to 16 carbon atoms) with known logP values (determination of the logP values by the retention times using linear interpolation between two successive alkanones).

Use examples:

Example A

5 **Phaedon larvae test**

Solvent: 7 parts by weight of dimethylformamide

Emulsifier: 2 parts by weight of alkylaryl polyglycol ether

10 To produce a suitable preparation of active compound, 1 part by weight of active compound is mixed with the stated amounts of solvent and emulsifier, and the concentrate is diluted with emulsifier-containing water to the desired concentration.

15 Cabbage leaves (*Brassica oleracea*) are treated by being dipped into the preparation of active compound of the desired concentration and are populated with larvae of the mustard beetle (*Phaedon cochleariae*) while the leaves are still moist.

20 After the desired period of time, the kill in % is determined. 100% means that all beetle larvae have been killed; 0% means that none of the beetle larvae have been killed.

In this test, at an exemplary active compound concentration of 100 ppm, the compound of the Preparation Example 1, for example, exhibits a kill of 100% after 7 days.

Example B

Plutella test

- 5 Solvent: 7 parts by weight of dimethylformamide
 Emulsifier: 2 parts by weight of alkylaryl polyglycol ether

10 To produce a suitable preparation of active compound, 1 part by weight of active compound is mixed with the stated amounts of solvent and emulsifier, and the concentrate is diluted with emulsifier-containing water to the desired concentration.

15 Cabbage leaves (*Brassica oleracea*) are treated by being dipped into the preparation of active compound of the desired concentration and are populated with caterpillars of the diamondback moth (*Plutella xylostella*) while the leaves are still moist.

After the desired period of time, the kill in % is determined. 100% means that all caterpillars have been killed; 0% means that none of the caterpillars have been killed.

20 In this test, at an exemplary active compound concentration of 100 ppm, the compound of the Preparation Example 1, for example, exhibits a kill of 100% after 7 days.

Example C

Spodoptera frugiperda test

- 5 Solvent: 7 parts by weight of dimethylformamide
 Emulsifier: 2 parts by weight of alkylaryl polyglycol ether

10 To produce a suitable preparation of active compound, 1 part by weight of active compound is mixed with the stated amounts of solvent and emulsifier, and the concentrate is diluted with emulsifier-containing water to the desired concentration.

15 Cabbage leaves (*Brassica oleracea*) are treated by being dipped into the preparation of active compound of the desired concentration and are populated with caterpillars of the armyworm (*Spodoptera frugiperda*) while the leaves are still moist.

After the desired period of time, the kill in % is determined. 100% means that all caterpillars have been killed; 0% means that none of the caterpillars have been killed.

20 In this test, at an exemplary active compound concentration of 100 ppm, the compound of the Preparation Example 1, for example, exhibits a kill of 100% after 7 days.

Example D

Heliothis armigera test

- 5 Solvent: 7 parts by weight of dimethylformamide
 Emulsifier: 2 parts by weight of alkylaryl polyglycol ether

10 To produce a suitable preparation of active compound, 1 part by weight of active compound is mixed with the stated amounts of solvent and emulsifier, and the concentrate is diluted with emulsifier-containing water to the desired concentration.

15 Soya bean shoots (Glycine max) are treated by being dipped into the preparation of active compound of the desired concentration and are populated with *Heliothis virescens* caterpillars while the leaves are still moist.

 After the desired period of time, the kill in % is determined. 100% means that all caterpillars have been killed; 0% means that none of the caterpillars have been killed.

20 In this test, at an exemplary active compound concentration of 100 ppm, the compound of the Preparation Example 1, for example, exhibits a kill of 100% after 7 days.

Example E

Diabrotica balteata test (larvae in soil)

Critical concentration test/soil insects – treatment of transgenic plants

5

Solvent: 7 parts by weight of dimethylformamide

Emulsifier: 1 part by weight of alkylaryl polyglycol ether

10

To produce a suitable preparation of active compound, 1 part by weight of active compound is mixed with the stated amount of solvent, the stated amount of emulsifier is added and the concentrate is diluted with water to the desired concentration.

15

The preparation of active compound is poured onto the soil. The concentration of the active compound in the preparation is virtually immaterial, only the amount by weight of active compound per volume unit of soil, which is stated in ppm (mg/l), matters. The soil is filled into 0.25 l pots, and these are allowed to stand at 20°C.

20

Immediately after the preparation, 5 pregerminated maize corns of the cultivar YIELD GUARD (trademark of Monsanto Comp., USA) are placed into each pot. After 2 days, the corresponding test insects are placed into the treated soil. After a further 7 days, the efficacy of the active compound is determined by counting the number of maize plants that have emerged (1 plant = 20% activity).

Example F

Heliothis virescens test (Treatment of transgenic plants)

- 5 Solvent: 7 parts by weight of dimethylformamide
Emulsifier: 1 part by weight of alkylaryl polyglycol ether

10 To produce a suitable preparation of active compound, 1 part by weight of active compound is mixed with the stated amount of solvent and the stated amount of emulsifier, and the concentrate is diluted with water to the desired concentration.

15 Soya bean shoots (Glycine max) of the cultivar Roundup Ready (trademark of Monsanto Comp. USA) are treated by being dipped into the preparation of active compound of the desired concentration and are populated with the tobacco budworm caterpillar *Heliothis virescens* while the leaves are still moist.

After the desired period of time, the kill in % is determined. 100% means that all caterpillars have been killed; 0% means that none of the caterpillars have been killed.